

**Case studies of aerosol retrievals over the ocean from multi-angle, multi-spectral photo-polarimetric remote sensing data**

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**Abstract.** To evaluate the global effects of aerosols on the direct radiative balance, tropospheric chemistry and cloud properties of the earth's atmosphere requires high-precision remote sensing that is sensitive to the aerosol optical thickness, size distribution, refractive index and number density. In this study we use the multi-angle 0.41, 0.55, 0.865 and 2.25  $\mu\text{m}$  channel data from the airborne Research Scanning Polarimeter to retrieve aerosol properties over the Pacific Ocean. We show that such photopolarimetric data are highly sensitive to the size distribution and refractive index of aerosol particles, which reduces the non-uniqueness in aerosol retrievals using such data as compared with less comprehensive data sets. Moreover, we find that polarized reflectances obtained at the shorter wavelengths (0.41 and 0.55  $\mu\text{m}$ ) are significantly less sensitive to the contribution of the ocean's upwelling light than total reflectance measurements, providing a natural tool for the separation between the estimation of oceanic and atmospheric scattering properties.

## 1. Introduction

Aerosols are believed to have a significant impact on the radiative balance and chemistry of the troposphere. Model computations show that the former effect, which includes the so-called indirect aerosol effect or aerosol-induced changes in the reflection properties of clouds, can be comparable in magnitude but opposite in sign to that of anthropogenic greenhouse gases [IPCC, 1996]. They also suggest that aerosol-induced heterogeneous reactions reduce the global average  $\text{NO}_x$  burden by half [Denenter and Crutzen, 1993]. Reliable evaluation of both of these effects and of aerosol transport models used to provide present and future aerosol climatologies, requires precise global measurements of the aerosol optical thickness, chemical composition, size distribution, and number density. Thus far, the only operational aerosol satellite product

available has been the optical thickness retrieved over the ocean using channel-1 radiances from the Advanced Very High resolution Radiometer (AVHRR) [Stowe and Ignatov, 1997]. The accuracy of this product is limited by the use of a single-wavelength ( $\lambda$ ), single-viewing angle ( $\theta$ ) algorithm, which necessitates making prior assumptions about all the other parameters of the atmosphere-ocean system. Although it is possible to improve the accuracy of the optical thickness retrievals from AVHRR measurements and also to determine the aerosol Ångström exponent by including channel-2 radiances [Higurashi and Nakajima, 1999] there are still many untested prior assumptions required in the retrieval process [Mishchenko *et al.*, 1999].

Aerosols also represent a problem for the space-borne remote sensing of ocean color by instruments such as the Coastal Zone Color Scanner (CZCS), the Sea Wide Field Scanner (SeaWiFS), and the Ocean Color and Temperature Scanner (OCTS) instruments. Ocean color monitoring uses estimates of ocean water-leaving radiances to derive information on phytoplankton concentrations. These radiances become largest in the visible part of the spectrum where the absorption by pure ocean water is least, but still represent less than 20% of the total radiance observed from space. To distinguish the ocean signal from atmospheric scattering, it is common to estimate the latter contributions from near-infrared ( $\geq 0.7 \mu\text{m}$ ) radiances which are least sensitive to water-leaving radiances (Gordon, 1997). Obviously any errors in the atmospheric correction (the estimation of aerosol composition, optical depth and size distribution) will be propagated into errors in ocean color and chlorophyll-like pigment concentration estimates. Moreover, any uncertainties in the separation of the observed radiances into the contributions from ocean and atmosphere will impose limits on the potential of shorter wavelength measurements to constrain aerosol retrievals.

An array of new satellite instruments has recently been deployed to provide more complete observations of the Earth's land, ocean, and atmosphere. We note in particular the MODerate resolution Imaging Spectrometer (MODIS) [King *et al.*, 1992], the Multiangle Imaging Spectro-Radiometer (MISR) [Diner *et al.*, 1991], and the POLarization and Directionality of the Earth's Reflectance (POLDER) instrument [Goloub *et al.*, 1991]. These instruments monitor the intensity (i.e., Stokes parameter  $I$ ) of sunlight reflected by terrestrial systems in more wavelengths (MODIS, MISR, POLDER) and from more viewing angles (MISR, POLDER) than the AVHRR instrument (Table 1), and in addition measure the linearly polarized component (i.e., Stokes parameters  $Q$  and  $U$ ) of reflected radiation (POLDER). The improvements that these instruments will make should, however, be contrasted with the accuracy requirement for climatologically useful aerosol retrievals [Hansen *et al.*, 1995]. Although the increased spectral range available from these instruments enhances their capability to retrieve particle size distributions, simulations performed for MODIS-type retrievals have already demonstrated significant difficulties in distinguishing between monomodal and multimodal size distributions [Tanré *et al.*, 1996]. The inclusion of measurements of the angular variation in the reflected radiation field in remote sensing allows for a higher sensitivity to aerosol type, but sensitivity analyses for MISR-type retrievals [Mishchenko and Travis, 1997] show that such data may still lead to ambiguous solutions for the aerosol refractive index and effective radius, both of which vary with relative humidity.

The fact that these intensity-only approaches to the retrieval of aerosol properties have such serious limitations suggests that an alternative measurement approach is necessary. Reflected solar radiation is, in general, polarized and contains embedded information about the intrinsic nature of aerosol particles as well as the underlying surface. Most of the detailed

physical information about aerosols (e.g., particle size distribution, composition based on the spectral signature of refractive indices, particle shape) is only readily available through the measurement and analysis of the spectral and angular polarization signature of the reflected radiation. The principal reason for the greater effectiveness of remote sensing by means of polarization measurements is the significantly greater sensitivity of polarization features to particle size, shape and refractive index as a function of scattering angle and wavelength than is the case for intensity measurements. When multiple scattering effects are included, the strength of intensity features is considerably diminished, while the single scattering information is largely retained in the polarization measurements.

Not only do polarimetric measurements contain more information about aerosols than intensity measurements, but there are also instrumental design and calibration advantages to making polarization measurements. For example, it is possible with a well designed polarimetric instrument to achieve absolute polarimetric accuracy of 0.2% on an operational basis. This high polarimetric accuracy ensures that any error in the absolute calibration of the polarized reflectance measurements used in this paper is almost identical to, and in the same sense as, the absolute calibration of the reflectance measurements.

An example of the sensitivity of polarization features to refractive index is given in Fig. 1, which shows the total and polarized intensity as a function of scattering angle for light with a wavelength of 0.865  $\mu\text{m}$  scattered once by particles with identical size distributions (effective radius of 1.0  $\mu\text{m}$ ) but different refractive indices  $m_\lambda$ . Another example, based on actual POLDER data, for a case where there is significant multiple scattering is given by Bréon and Goloub (1998) who compare the sensitivity of polarized and total reflectances to cloud droplet effective radii. In spite of the potential of polarimetric measurements to retrieve aerosol properties,

comparisons with ground-based sun-photometer measurements still reveal systematic errors in POLDER-derived Ångström exponents [Goloub *et al.*, 1999]. Recent analyses suggest that the simultaneous occurrence of small and large aerosol particles may be responsible for some of the discrepancies found in these retrievals, which currently assume monomodal size distributions [Deuzé *et al.*, 2000]. Based on in-situ sampling measurements and our understanding of the processes of aerosol formation, removal and transport [Jaenicke, 1993], such bimodal aerosol mixtures are expected to be ubiquitous, but the resolution of the two modes in the aerosol size distribution has proved difficult with the relatively small useful spectral range (0.67 $\mu\text{m}$  and 0.865 $\mu\text{m}$ ) of POLDER observations.

In this paper, we discuss aerosol retrievals over the ocean using data collected with the Research Scanning Polarimeter (RSP) instrument [Cairns *et al.*, 1999], an airborne instrument that is functionally similar to the Earth Observing Scanning Polarimeter (EOSP) [Travis, 1993]. The RSP instrument was designed to provide high-precision, multi-angle measurements of polarization and intensity in a wide spectral range from the visible to the short-wave infrared (Table 1). This allows us to build on the analyses and results of the MODIS, MISR and POLDER teams, since the RSP instrument provides measurements that are similar to, and in many respects more comprehensive than, all three satellite instruments. In a recent paper, we demonstrated by applying inversion methods to different subsets of RSP data how the range of available measurements in a data set affects the non-uniqueness problem faced in the parameter estimation problem which is the remote sensing of aerosols (Chowdhary *et al.*, 2001). Our results based on employing just two narrow-band channels (0.865 and 2.25  $\mu\text{m}$ ) demonstrated the superiority of using multi-angle intensity **and** polarization measurements over approaches that use more limited measurement sets. In this paper we explore the use of observations at

shorter wavelengths (0.41 and 0.55  $\mu\text{m}$ ) and examine the water-leaving contribution to the observed intensity and polarization. This analysis is designed to determine whether the signals from the ocean and from aerosols at these shorter wavelengths can be better separated by using both intensity and polarization measurements. We also examine, using different subsets of RSP data, how the completeness of the available data set affects aerosol retrievals and the consequences of any errors in these retrievals for the estimation of ocean color.

## **2. Instrument design and performance**

The properties of the RSP instrument and measurements that form the basis for the analyses in the next section are discussed in the following. The 14 mrad instantaneous fields of view (IFOV) of the six boresighted RSP telescopes are continuously scanned by a polarization-neutral two-mirror system which allows 152 viewing-angle samples to be acquired over a  $120^\circ$  swath ( $\pm 60^\circ$  from nadir). The desired polarization-insensitive scanning function of the RSP is achieved by the use of a two-mirror system with the mirrors oriented such that any polarization introduced at the first reflection is compensated for by the second reflection. The refractive telescopes are paired, with each pair making measurements in three spectral bands. One telescope in each pair makes simultaneous measurements of the linear polarization components of the intensity in orthogonal planes at  $0^\circ$  and  $90^\circ$  to the meridional plane of the instrument (using Wollaston prisms to spatially separate the orthogonal polarizations onto a pairs of detectors) while the other telescope in a pair simultaneously measures equivalent intensities in orthogonal planes at  $45^\circ$  and  $135^\circ$ . These measurements in each instantaneous field of view in a scan provide the simultaneous determination of the Stokes parameters  $I$ ,  $Q$ , and  $U$  in all nine spectral bands with a wide dynamic range (14-bit digitalization) and high signal-to-noise ratio

(2000 at a Lambertian equivalent reflectance of 0.3). This approach ensures that the polarization signal is not contaminated by scene intensity variations during the course of the polarization measurements, which could create “false” or “scene” polarization.

The instrument has nine spectral channels that are divided into two groups based on the type of detector used: visible/near infrared (VNIR) bands using UV-enhanced silicon photodiodes at wavelengths of 410 (30), 470 (20), 550 (20), 670 (20), 865 (20) and 960 (20) nm and short-wave infrared (SWIR) bands using HgCdTe photodiodes (cooled to 163K) at wavelengths of 1590 (60), 1880 (90), and 2250 (120) nm. Dichroic beam splitters are used for spectral selection, while interference filters define the spectral bandpasses of each band. The parenthetic figures are the full width at half maximum (FWHM) bandwidths of the spectral bands. These spectral bands sample the spectrum of reflected solar radiation over most of the radiatively significant range, with measurements under typical clear sky conditions ranging from significant Rayleigh scattering (410nm) to single scattering by aerosol (2250nm) within a single data set.

The expected radiometric uncertainty is 3.5% using a reflectance-based mountaintop calibration. The uncertainty in degree of linear polarization measurements is 0.2% [Cairns *et al.*, 1999], based on instrument characterization and pre- and post- flight relative calibrations of the detectors for a particular spectral band within a given telescope. Since the polarimetric and radiometric calibration paths are independent this implies that the polarized reflectance measurements have an uncertainty that is almost identical to the radiometric uncertainty (i.e. 3.51%).

### **3. Model description and data simulations**



We adopt for our analyses the same simple atmosphere-ocean model system used in Chowdhary *et al.* (2001), see Fig. 2. Aerosol particles are homogeneously mixed with molecules below the aircraft and are generally assumed to have either a monomodal (accumulation) or a bimodal (accumulation and coarse mode) log-normal size distribution [cf. Nakajima *et al.*, 1996]. Trimodal (and higher number mode) solutions are also possible and the retrievals presented below are therefore by no means unique. However a bimodal solution is the most parsimonious solution that is consistent with both the prior information from *in situ* sampling measurements and the remote sensing data used in this study. Following Hansen and Travis [1974], we use parameters that can be retrieved from remote sensing data to characterize each aerosol mode, i.e., the effective radius ( $r_{\text{eff}}$ ) and effective variance ( $v_{\text{eff}}$ ) of its size distribution, its refractive index ( $m_\lambda$ ) as a function of wavelength, and its optical thickness ( $\tau_\lambda$ ) as a function of wavelength. In addition, we derive from these optical parameters the column number density of particles ( $N$ ) for each aerosol component [Mishchenko *et al.*, 1997], a physical quantity of interest to the evaluation of the indirect aerosol effect. When retrieving the refractive index we require its real part to be decreasing as the wavelength increases in the near infrared part of the spectrum, and to remain constant in the visible part of the spectrum. Such spectral behavior of ( $m_\lambda$ ) is consistent with trends found in laboratory and *in-situ* measurements of aerosol properties (e.g., D'Almeida *et al.*, 1991), and facilitates the inversion process by reducing the size of the aerosol parameter space. In those cases where the absorption optical depth of aerosols is small, i.e. low aerosol optical depths and/or single scatter albedo close to unity, reflectance measurements over the ocean are not very sensitive to absorption. Also, scattering matrices of aerosol particles are not very sensitive to the imaginary part of refractive index except for strongly absorbing materials. For simplicity we therefore set the imaginary part of the refractive index to zero, i.e., we ignore

aerosol absorption. This indicates that there is a non-uniqueness issue in the retrieval of aerosol single-scatter albedo from reflectance and polarized reflectance measurements for low aerosol optical depths [Mishchenko and Travis 1997b], unless sunglint measurements are included in the analysis. The single scattering properties of aerosol particles are obtained using the Mie-theory, i.e., the implicit assumption here is that the particles are spherical and homogeneously mixed. If the sphericity assumption is not true, then it will not be possible to achieve a satisfactory fit to the polarization measurements [Mishchenko *et al.*, 1997a]. In such a case (e.g., coarse mode consists of dust particles) we would then use computations of light scattering by shape distributions of polydisperse spheroids to represent non-spherical particles. As regards the homogeneity assumption, in all the extant data that we have analyzed Mie theory calculations have provided excellent fits. Thus, it appears that the retrieved refractive indices are at least representative of an effective medium approximation [Chylek *et al.* 2000].

Gaseous absorption is assumed to occur above the scattering atmosphere, in the stratosphere, and is simply modeled as an attenuation of the direct beam of the sun. This assumption is suitable for absorption by ozone in the Chappuis band, and we use for the latter absorption an ozone amount of 272 Dobson Units (DU) which is consistent with both TOMS and multifilter rotating shadowband radiometer (MFRSR) measurements. Our assumption for gaseous absorption is probably less realistic for NO<sub>2</sub>; however, the modeled amount ( $5 \pm 1.5$  ppb from MFRSR measurements, Alexandrov *et al.* 2001) is quite plausible given our current understanding of atmospheric NO<sub>2</sub> concentrations and distributions [Shaw, 1976, Noxon, 1978, Pujadas *et al.* 2000]. It should be noted that the effects of gaseous absorption are corrected for in the RSP data, rather than being included in the model. The model comparisons with RSP data presented below are therefore for a purely scattering atmosphere. The molecular optical

thickness that we use for the scattering atmosphere is that given by Hansen and Travis (1974) with a scale-height of 8 km, which defines the amount of Rayleigh scattering above and below the aircraft.

The reflection and transmission properties of the ocean surface are calculated using the geometrical optics approach with a wind-speed-dependent surface slope distribution [Cox and Munk, 1954] and with surface shadowing effects [Sancer, 1969] and scattering by oceanic foam [Koepke, 1984] taken into account. When inverting visible remote sensing data obtained over ocean bodies, one commonly approximates the intensity of the upwelling underwater light by means of a Lambertian surface with an albedo empirically related to the chlorophyll-like pigment concentration  $[C]$ , in  $\text{mg/m}^3$  [Gordon and Morel, 1983]. Such approximations are valid for retrievals based on near-nadir single-viewing angles and photometric measurements. However, the sensitivity of RSP measurements to the bidirectional [Morel and Gentili, 1995] and especially polarized nature [Voss and Fry, 1984] of underwater light scattering requires a more rigorous approach for both the simulation and inversion of multiangle polarimetric remote sensing data of atmosphere-ocean systems. As an ansatz towards solving these problems, we perform polarized radiative transfer computations for the underwater light using realistic bulk oceanic scattering properties and perform sensitivity studies for the dataset analyzed in the present work. We assume for this purpose that scattering in the ocean body is well approximated by a homogeneous mixture of clean oceanic water and hydrosol particles (Fig. 3). Scattering by clean oceanic water is modeled as Rayleigh scattering with a depolarization factor of 0.09 [Morel, 1974], and with the scattering and absorption coefficients given by Hale and Querry (1973). The scattering by hydrosol particles is based on the application of Mie theory to particles with an inverse fourth power law size distribution [Stramski and Kiefer, 1991], lower

bound of  $0.001\ \mu\text{m}$  and an upper bound of  $3.0\ \mu\text{m}$ , and a refractive index of  $1.10-0.0025i$  relative to that of water. This microphysical model gives rise to a phase function  $F_{11}$  (Fig. 3a) that closely resembles the averaged one given by Mobley *et al.* (1993) for all the hydrosol phase functions measured by Petzold (1977). Moreover, the linear polarization ratio  $-F_{21}/F_{11}$  for these particles is similar to that found in laboratory measurements of phytoplankton [Volten *et al.*, 1998], and is consistent with the rather small variation in the degree of linear polarization (Fig. 3b) found for bulk oceanic waters [e.g., Kadyshevich and Lyubovtseva, 1973]. The scattering and absorption coefficients for hydrosol scattering are taken from Morel (1988) for Case-1 water, i.e., oceanic surface waters for which the optical properties are dominated by phytoplankton and their by-products. Note that the dependence of these coefficients on  $[C]$  is consistent with that used for Lambertian ocean models. Finally, we adopt a bulk optical thickness of 10 for the ocean body.

Multiple scattering computations for the transport of light through this model atmosphere and ocean are performed using the fast doubling/adding method [De Haan *et al.*, 1987; Chowdhary, 1999]. Different sets of Gaussian quadrature points are used for the skylight and underwater light directions to increase computational efficiency, and we renormalize the ocean surface scattering properties to account for multiple reflections between ocean waves (isotropic renormalization) and to ensure conservation of energy (refraction peak renormalization). Although the numerical precision of our computations is better than 5 significant figures, uncertainties in the values, or spectral dependence of some of the parameters adopted for our atmosphere-ocean model lead to a lower confidence in the verisimilitude of our model when comparisons are being made to measurements of an actual ocean-atmosphere system. The principal sources of uncertainty are the surface-fraction [Monahan and O'Muircheartaigh, 1980]

and spectral albedo [Frouin *et al.*, 1996] of oceanic foam, the wind-directionality of the sun glint [Masuda, 1988], and the scattering coefficient for hydrosols (Loisel and Morel, 1998]. We adopt an uncertainty of 5% for all our radiative transfer results to account for the potential of such errors in *a-priori* assumptions. This uncertainty in the modeling of the system is also the reason that reflectance and polarized reflectance measurements are used in the following analyses, since it is easier to identify and diagnose modeling errors and uncertainties than in the (more accurate) degree of linear polarization.

## 4. Measurements and aerosol retrievals

### 4.1 Data set

The data used for the present study were acquired off the coast of Santa Cruz Is., California on October 14, 1999 with the RSP instrument mounted on an aircraft and the scanner oriented so that the plane of the scan is directed along the direction of the aircraft ground track. The scattering geometry for this data set is given by the solar zenith angle  $\theta_0 = 43^\circ$  and azimuth difference  $\phi - \phi_0 = 3^\circ$  between the principal plane and RSP scan plane. The altitude (3000 m) and speed (50 m/s) of the aircraft were chosen such that successive nadir views were one IFOV apart and the same point at the ground was thus seen from multiple viewing angles, with the nadir pixel size being 42 m. Vignetting of the RSP scan by the skin of the aircraft made data at viewing angles larger than  $\pm 40^\circ$  from nadir unavailable. In what follows, we actually show and analyze for each viewing angle  $\theta$  the average of 40 consecutive same-viewing angle measurements. Our study focuses on measurements obtained at 0.41, 0.55, 0.865, and 2.25  $\mu\text{m}$ . The longer two wavelengths are used to distinguish fine and coarse mode aerosol properties. Measurements obtained between these two wavelengths can be used to retrieve the spectral

variation of refractive indices between 0.865 and 2.25  $\mu\text{m}$ . The shorter two wavelengths are chosen to study the contributions of polarized and total reflectances that originate from underwater light scattering, and to assess the retrieval of aerosol properties from polarized visible light. These four spectral bands used were chosen as exemplars of the effects of atmospheric and ocean contributions and because they span the full spectral range of the RSP measurements. The bands at 0.47 and 0.67  $\mu\text{m}$  were excluded from the analysis for simplicity and in the case of the 0.67  $\mu\text{m}$  band because it can contain poorly characterized contributions from ocean body scattering [Toole *et al.* 2000]. The remaining bands are not included in the analysis presented below because they are used for water vapor estimates (0.96  $\mu\text{m}$ ), cirrus clouds screening (1.88  $\mu\text{m}$ ) or were not functional (1.59  $\mu\text{m}$ ).

A Multi Filter Rotating Shadowband Radiometer (MFRSR) instrument was also used during the period of the RSP data collection to provide estimates of the aerosol optical depth, effective radius, effective variance,  $\text{NO}_2$  and ozone column abundances and the downwelling horizontal irradiance in five narrow spectral bands at 0.415, 0.501, 0.616, 0.672 and 0.869  $\mu\text{m}$  [Alexandrov *et al.* 2001]. The MFRSR was located 1km inland near Oxnard, CA and airborne RSP measurements of the surface around the MFRSR were used to characterize the Bidirectional Reflectance Distribution Function (BRDF) of the surface so that the method of King *et al.* [1979] could be used to estimate the single scatter albedo of the aerosols. The measured aerosol optical depth at 0.55  $\mu\text{m}$  was 0.14 ( $\pm 0.01$ ) and the single scatter albedo was estimated to be 0.95 ( $\pm 0.02$ ) with negligible spectral variation. The absorption optical depth was therefore less than 0.01, which is why the neglect of absorption for the measurements presented below is considered reasonable.

#### 4.2 Aerosol retrieval using the full set of RSP data

Our bimodal aerosol retrieval method incorporates the two steps followed in Chowdhary *et al.* (2001). The first step consists of using the 2.25  $\mu\text{m}$  total and polarized reflectance measurements to estimate the coarse mode aerosol parameters under the assumption that the accumulation mode aerosol optical thickness is (negligibly) small at this wavelength. The second step consists of using this estimate, together with the 0.865  $\mu\text{m}$  total and polarized reflectance measurements, to estimate the parameters of the accumulation mode aerosol. In addition, we use in the present work the polarized reflectance measurements at 0.41 and 0.55  $\mu\text{m}$  to verify, or refine, our aerosol estimates under the assumption that the ocean body is black (i.e., ignoring the contribution of water-leaving radiances). This process is iterated until it converges to a solution that is consistent with all of the above data. The rationale for using the visible polarized reflectances to constrain aerosol properties is discussed below, while in Sec. 4.3 we address the contribution of water-leaving radiances.

The upper panels of Fig. 4 show data and the type of results that are obtained using this iterative estimate of aerosol properties. The left and right panel are for the total reflectance  $\pi I(\mu_0 S)^{-1}$  and for the linearly polarized reflectance  $\pi(Q^2 + U^2)^{1/2}(\mu_0 S)^{-1}$  as a function of the view zenith angle, respectively. The view zenith angle  $\theta$  is positive in the direction of the plane's flight (see Fig. 2), while  $\mu_0 = \cos\theta_0$  and  $S$  is the extra-terrestrial solar flux. Hence, the sharp increase in reflectance seen for positive viewing angles corresponds to the sun glint profile. The mean and standard deviation of the RSP measurements are shown by the lines and error-bars, respectively. The symbols show model simulations for the estimated bimodal aerosol model (hereafter referred to as bimo-1; see Table 2). The goodness of the fit between measurements and

simulations is given for each wavelength in the upper left corner of the panels by the normalized chi-square value ( $\chi^2$ ) defined as

$$\chi^2 = \frac{1}{N} \sum_{i=1}^N \frac{(y_i - x_i)^2}{S_i} \quad (1)$$

where  $N$  is the number of viewing angles analyzed,  $y_i$  is the simulated reflectance for viewing angle  $i$ ,  $x_i$  is the average reflectance for viewing angle  $i$ , and  $S_i$  is the standard deviation of the combined error sources for viewing angle  $i$ , viz.

$$S_i = \sqrt{(0.05 y_i)^2 + (0.035 x_i)^2 + \sigma_i^2}. \quad (2)$$

In Eq. (2),  $\sigma_i$  is the standard deviation of the measurements for viewing angle  $i$ , i.e., it is given by

$$\sigma_i = \sqrt{\frac{\sum_{j=1}^P (x_i^j - x_i)^2}{P-1}} \quad (3)$$

where  $P$  is the number of pixels used for each viewing angle analysis, and  $x_i^j$  is the reflectance of pixel  $j$  for viewing angle  $i$ . We used the standard deviation in the computation of the combined error rather than the standard error because pixel-to-pixel variations are caused not only by instrumental noise, but also by reflectance variations in the ocean surface and aircraft orientational changes. Equation (1) can therefore only be regarded as an *ad hoc* measure for the agreement between measurements and model calculations, with  $\chi^2 \leq 2$  providing a qualitative indication of acceptable results. Note also that angular variation of the reflected radiances is smooth, suggesting the existence of strong correlations between different-viewing angle measurements that should be accounted for in more rigorous statistical measures [Dubovik and King, 2000]. In what follows, we use only the angular range of  $-35^\circ \leq \theta \leq 20^\circ$  to compute



normalized chi-square values in order to avoid serious contamination by the sun glint, and we exclude polarized reflectance measurements smaller than  $5 \cdot 10^{-5}$  (horizontal dashed line) from this computation since they are at the digitization level of the RSP measurements.

There are a number of points to be noted for Figs. 4*a* and 4*b*. Firstly, the sun glint angular profile is quite sensitive to the wind-speed  $W$ , which allowed us to constrain the wind speed to 4.5 ( $\pm 0.5$ ) m/s. Secondly, the retrieved aerosol model has an optical thickness at a reference wavelength of 0.55  $\mu\text{m}$  of 0.148 (Table 2) which is in good agreement with the value of 0.14 measured that day at the same time at Oxnard, CA, using an MFRSR. Thirdly, the total reflectance measured at 0.41 and 0.55  $\mu\text{m}$  is significantly larger than that which is simulated in these bands. The differences can be attributed to the neglect of water-leaving radiances in our computations (Sec. 4.3), although the variation of water-leaving radiances with the pigment concentration in the ocean makes it difficult to verify such results without complimentary knowledge on the biotic state of the ocean. The mixing of atmospheric and ocean contributions to the total upwelling reflectance observed at these shorter wavelengths may in fact pose a problem for the retrieval of accumulation-mode aerosol properties if one uses only photometric measurements. This is because the effective size parameter,  $(2\pi r_{\text{eff}})/\lambda$ , of sub-micron particles only becomes large enough at visible wavelengths for their single-scattering properties to be well differentiated from Rayleigh scattering (*cf.* Mishchenko and Travis, 1997). On the other hand, polarized reflectances measured in the visible part of the spectrum do provide useful constraints on the accumulation mode aerosol properties. Consider for example in Figs. 4*c* and 4*d* the results of an aerosol retrieval in which the polarized reflectance measurements at 0.41 and 0.55  $\mu\text{m}$  are ignored. Note that a different bimodal aerosol model, hereafter referred to as bimo-2, whose accumulation mode component differs from the one of bimo-1 by as much as 0.07 in

refractive index (Table 2), is just as consistent with reflectance and polarized reflectance measurements at 0.865 and 2.25  $\mu\text{m}$  as the bimo-1 model. It is only by including the 0.41 and 0.55  $\mu\text{m}$  polarized reflectance that we start to observe significant discrepancies between measured and simulated polarized reflectances. We show in the next section that such discrepancies can *not* be bridged by the inclusion of water-leaving radiances.

#### *4.3 Contribution of waterleaving radiances*

To evaluate the contribution of waterleaving radiances to the total and polarized reflectances observed by the RSP instrument, we perform here sensitivity studies for various ocean models. Consider first the case of a purely molecular scattering ocean in Figs. 5*a* and 5*b* (blue dashed lines). Note that the total reflectance computed at 0.41  $\mu\text{m}$  is now larger than the corresponding RSP measurements, while the total reflectance computed at 0.55  $\mu\text{m}$  is still less than the observed reflectance. This result is consistent with what one would expect for an ocean body with appreciable amounts of chlorophyll-like pigments which absorb strongly in the blue but weakly in the green part of the visible spectrum [Morel, 1988, and references therein]. By subsequently increasing the amount of Chlorophyll-like pigments in our ocean model we observe a convergence between our numerical calculations (solid grey lines) and the RSP measurements. Thus, it is possible to retrieve an aerosol model and an ocean biophysical model that is consistent with all of the multi-spectral, multi-angle polarized RSP measurements. Furthermore, comparing Figs. 5*a* and 5*b* shows that the polarized reflectance is a factor of two less sensitive to the contribution from underwater light scattering than the reflectance. Indeed, the difference between a purely molecular ocean (blue dashed line) and one with  $[C] = 1 \text{ mg/m}^3$  (upper bound for most of the ocean, solid grey line) is now about equal to the uncertainty adopted for our simulations.

The weak sensitivity of polarized reflectances to ocean color justifies their inclusion in our retrieval method, and shows the potential of polarimetry to separate ocean color monitoring from atmospheric correction. We illustrate the estimation of an ocean biophysical model from measurements of this type by inverting the residual total reflectances to obtain the chlorophyll-like pigment concentration  $[C]$ . The residual reflectances for our bimodal aerosol fit, shown in Fig. 5c, only become significant for measurements at wavelengths shorter than  $0.670\text{ }\mu\text{m}$ , which is spectrally consistent with Case-1 water-leaving radiances [Gordon, 1997]. Approximating these radiances away from the sunglint by radiances originating from a Lambertian ocean leads to ocean body albedos of 0.0275, 0.024 and 0.0097 at 0.41, 0.47 and  $0.55\text{ }\mu\text{m}$ , respectively (Fig. 5d). The latter albedos are consistent with a Chlorophyll-like pigment concentration  $[C]$  of  $0.30\text{ mg/m}^3$  based on the semi-empirical curves shown in Fig. 5e [cf. Morel, 1988], which is not unusual for this region of the ocean.

#### *4.4 Aerosol retrieval using sub-sets of RSP data*

We now examine how the accuracy of aerosol retrievals is affected by limiting the analysis to subsets of data. The rationale for this investigation is to examine what aspects of the multivariate aerosol model different measurement sets are sensitive to, with a view of understanding the types of discrepancies one might expect from retrievals using different satellite instruments. Figures 6a and 6b show the results of a bimodal aerosol fit that uses only the multi-angle, multi-spectral total reflectance data subset. These measurements have a spectral range ( $0.41\text{ }\mu\text{m}$  to  $2.25\text{ }\mu\text{m}$ ) that is similar to the one monitored by MODIS. Their multi-angle nature is also comparable to that of MISR data, i.e., while their angular range is smaller they were obtained near the solar principal plane to maximize the range of scattering angles observed.

Figure 6a shows that the observations at 0.865  $\mu\text{m}$  and 2.25  $\mu\text{m}$  can be well reproduced using the bimodal set of the water soluble and oceanic mix of the bimo-S aerosol model specified in Table 3. Note that these components are the same as those adopted by WMO (1986) except for (i) limiting the width of the oceanic aerosol size distribution to avoid incorporating unrealistically large ‘phantom’ particles (Lacis and Mishchenko, 1995), and (ii) ignoring the strong absorption by the water soluble particles to compare with the results in Sec. 4.2. Pre-defined sets of aerosol candidate models such as these are commonly used for the inversion of photometric remote sensing data even though the temporal and spatial variability observed for tropospheric aerosols is much larger. This is because the information content provided by multi-angle, multi-spectral photometric remote sensing data is often insufficient to even partially resolve the multi-dimensional aerosol parameter space. Hence, the accuracy with which aerosols can be retrieved depends in these cases as much on the initial choice of aerosol candidate model as on the fit between measurements and simulations. Including polarization in the remote sensing of aerosols reduces the non-uniqueness of the inversion problem because the measurements are sensitive to refractive index and allow for the discrimination and quantification of two modes of the aerosol size distribution (see Fig. 6b). For example, the presence of the WMO (1986) water-soluble (oceanic) aerosol particles causes the polarized reflectance to decrease much more (less) than observed by the RSP instrument for  $\theta \approx -32^\circ$  and  $\lambda = 0.865 \mu\text{m}$  ( $\lambda = 2.25 \mu\text{m}$ ). In Figs. 6c and 6d we consider the results of a bimodal aerosol retrieval based on the inversion of multi-angle photo-polarimetric measurements obtained at  $\lambda \leq 0.865 \mu\text{m}$ , i.e., of observations that have the same spectral range as those obtained by the POLDER instrument. There are three features of our analysis of this data subset that should be emphasized. First, no solutions could be found using the water-soluble and/or oceanic aerosol models of Table 3 as substitutes for the

accumulation and/or coarse mode aerosol in our retrieval algorithm. Second, the accumulation mode component of the retrieved bimodal aerosol resembled that of bimo-1 in Table 2 which is consistent with our discussion of Figs. 4c and 4d. Third, the 0.865  $\mu\text{m}$  and 0.55  $\mu\text{m}$  polarized reflectance measurements remained sensitive enough to large particles to rule out many coarse mode aerosol models. Such results are encouraging, suggesting that the use of polarimetry remains a powerful remote sensing tool even for small spectral ranges. Nevertheless, the absence of 2.25  $\mu\text{m}$  measurements leads to the retrieval of a bimodal aerosol (hereafter referred to as bimo-3) whose coarse mode particles are smaller and have a smaller refractive index than those of the initial retrieval using all the RSP data (*cf.* bimo-1 in Table 2). This is because the 2.25  $\mu\text{m}$  measurements are essentially insensitive to accumulation mode aerosols but provide much of the sensitivity to coarse mode aerosols.

An alternative to the bimodal aerosol candidate models examined above, is to use monomodal aerosol models when analyzing sub-sets of RSP-like data. This approach is less time consuming since the dimensionality of the parameter space has been reduced and the non-uniqueness problems discussed above are buried in the *a priori* assumption about the functional form of the aerosol size distribution. Nonetheless, if such distributions provide acceptable results their simplicity is a desirable feature in the estimation problem which constitutes an aerosol retrieval. A monomodal model is not however acceptable for the analysis of the complete RSP data set and so no monomodal inversion on the complete RSP data set is presented. Figures 7a and 7b show the results of such a retrieval using only the multi-angle, multi-spectral total reflectance data. In Fig. 7a it can be seen that the observations at 0.865  $\mu\text{m}$  and 2.25  $\mu\text{m}$  (solid lines) are now consistent with two different monomodal aerosol models (symbols). These two microphysical models, henceforth denoted mono-1 (triangles) and mono-2 (boxes), have the

same effective radius ( $0.4\ \mu\text{m}$ ), but differ significantly in refractive index (0.06, see Table 2). The polarized reflectance measurements (Fig. 7b) are completely inconsistent with these two microphysical models, ruling them out as acceptable solutions to the aerosol retrieval problem. This further demonstrates the sensitivity of polarization measurements to the particle refractive index as compared to reflectance measurements. Once again we observe that a radiance-only data set can lead to the retrieval of aerosol models that are significantly in error without any indication from the data itself that this is the case. In Figs. 7c and 7d we consider the results of a monomodal aerosol retrieval for a data set of multi-angle photo-polarimetric measurements obtained at  $\lambda \leq 0.865\ \mu\text{m}$ . The relatively large optical thickness of accumulation mode particles still allows for an acceptable retrieval using a monomodal aerosol model (hereafter denoted by mono-3) with a refractive index and size distribution close to those of the accumulation mode component of bimo-1, which was our original retrieval (Table 2). Note also that while the mono-3 aerosol optical thickness is significantly larger (34%) than that of the bimo-1 accumulation mode aerosol at  $0.55\ \mu\text{m}$ , its column particle number density ( $N$ ) is only slightly larger (5%) than that of the bimo-1 accumulation mode aerosol. Mishchenko *et al.* (1997b) found that it is much more difficult to determine  $N$  than to retrieve the optical depth for accumulation mode particles, based on single-channel sensitivity analyses. The mono-3 aerosol retrieval results suggests that this problem may well be reduced by including multi-spectral, multi-angle measurements of intensity *and* polarization in the inversion process

For the sake of completeness, we show in Fig. 8 the water-leaving residual reflectances based on aerosol retrievals assuming a monomodal aerosol size distribution, as described above. The results for the mono-2 aerosol model are given in Fig. 8a, where it is apparent that the atmospheric correction using intensity only measurements and an assumed monomodal size

distribution (based on the model shown in Fig. 7a) leads to significant errors in the bidirectional behavior of the ocean-body reflectance (Fig. 8b). The residual water-leaving reflectances for the mono-3 aerosol model are shown in Fig. 8c. The bidirectional behavior of the ocean-body reflectance that is inferred for this case (Fig. 8d) is similar to the results shown in Fig. 5d. The fact that the atmospheric correction using POLDER-like measurements yields better results for the ocean-body analyses than reflectance-only measurements is provocative, but any definitive conclusions about the relative superiority of these two types of measurement will require a more detailed and complete analysis.

## 5. Conclusions

An analysis of data obtained over the ocean by an airborne version of EOSP, the research Scanning Polarimeter (RSP), shows that bimodal size information, composition (via refractive index), aerosol optical depth and number density can be retrieved from multi-spectral, multiangle polarization measurements. The aerosol optical depth retrieved from the RSP data is consistent with sun-photometer measurements (difference of 0.008 at 0.55  $\mu\text{m}$ ). The weak sensitivity of polarized reflectances at shorter wavelengths (0.41 and 0.55  $\mu\text{m}$ ) to ocean color allows for their inclusion in the retrieval of aerosol properties and shows the potential of polarimetry to separate ocean color monitoring from atmospheric correction. We note that analyses of reflectance-only data subsets are not capable of distinguishing monomodal from bimodal aerosol solutions, nor do they provide strong constraints on aerosol refractive indices. The errors in aerosol retrievals from these reflectance-only data subsets are seen to propagate into errors in the estimated ocean color. The simulated examples of POLDER retrievals indicates useful sensitivity to the refractive index, size and optical depth of the accumulation mode aerosol, but the absence of longer

wavelength (2.25  $\mu\text{m}$ ) measurements limits the amount of information that can be obtained about the coarse mode aerosol. Since the errors in the estimated accumulation mode are small the estimate of ocean color from the POLDER-like data subset is similar to that obtained from the complete data set, suggesting that the inclusion of polarized reflectances from the shorter wavelength bands of POLDER may be useful in improving aerosol retrievals from this type of data.

The accurate polarimetric measurements made by the RSP, over a broad angular and spectral range, significantly reduce the non-uniqueness problem in the retrieval of aerosol properties by providing information about the spectral refractive index and both accumulation and coarse mode size distribution parameters. This capability allows aerosol parameters to be determined with the accuracy needed for long-term monitoring of the direct and indirect aerosol forcings of climate. The analysis of polarized reflectances at shorter wavelengths (0.41 and 0.55  $\mu\text{m}$ ) indicates the potential of these measurements to further improve aerosol retrievals and atmospheric correction for ocean color monitoring.

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## Figures

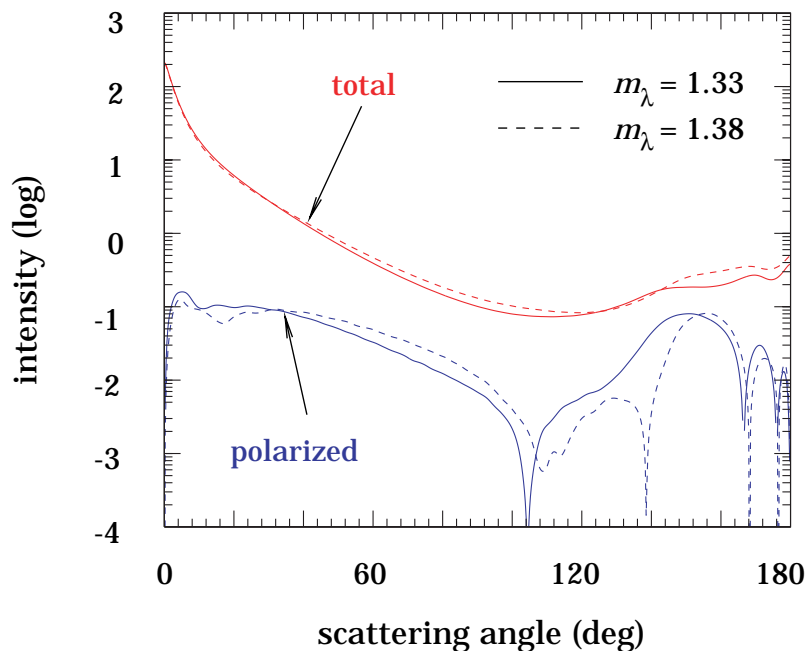


Figure 1. The total and polarized intensity of light scattered once at the wavelength  $\lambda = 0.865 \mu\text{m}$  as a function of scattering angle by a particle with effective radius  $r_{\text{eff}} = 1.0 \mu\text{m}$  and refractive index  $m_{\lambda} = 1.33$  (solid lines) and 1.38 (dashed lines).

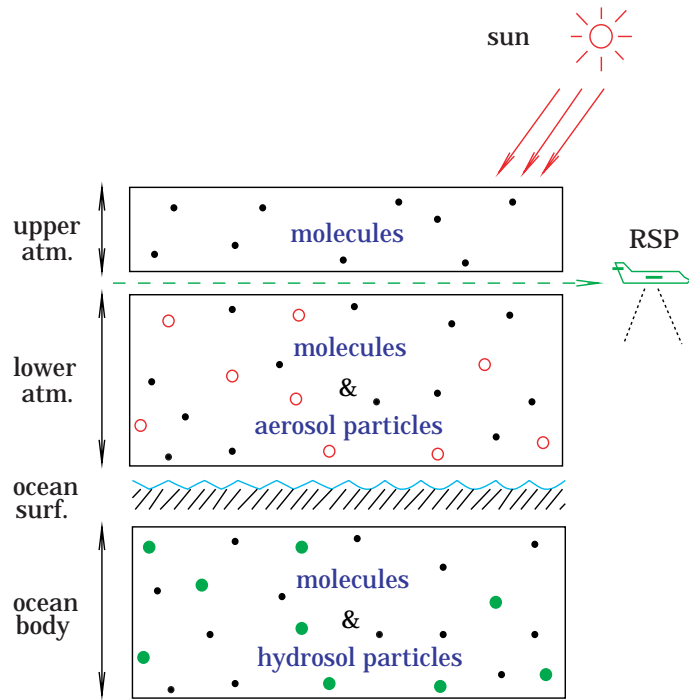


Figure 2. The atmosphere - ocean system used for simulating RSP data sets obtained from aircraft over the ocean.

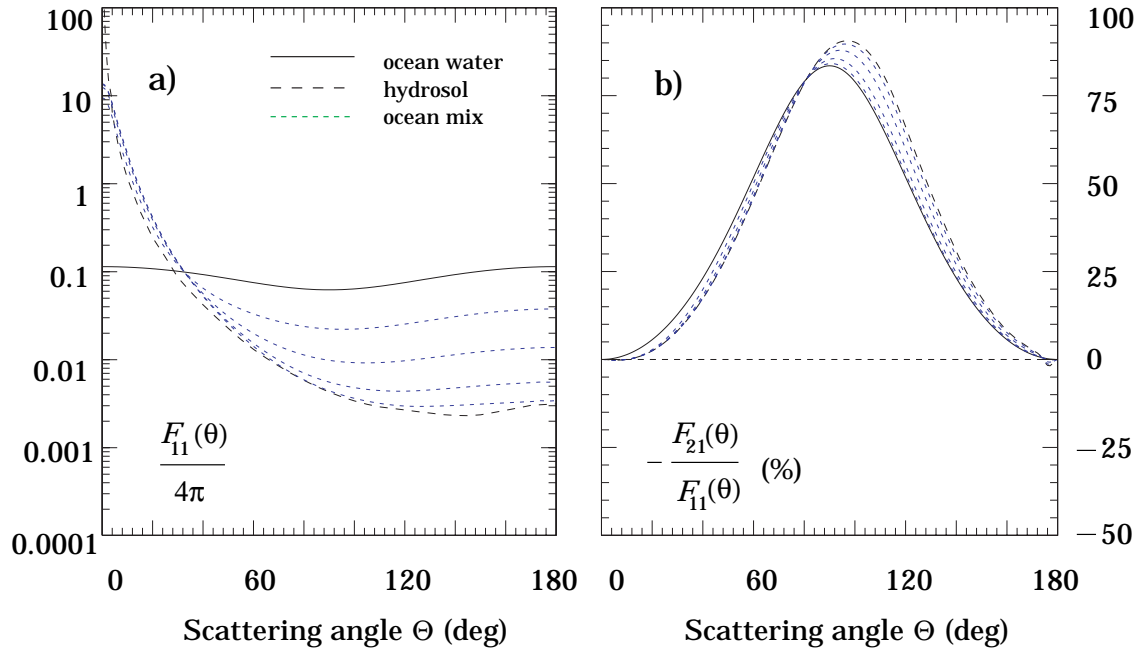


Figure 3. The phase functions  $F_{11}$  normalized by  $4\pi$  (left) and the linear polarization ratios  $-F_{21}/F_{11}$  (right) used for the underwater light computations as a function of the scattering angle. The hydrosol in the left and right panel stands for the average values used by Mobley *et al.* (1993) and the hydrosol particles specified in the text, respectively. The ocean mix curves are shown for chlorophyll-like pigment concentrations  $[C] = 1.000, 0.100, 0.010, \text{ and } 0.001 \text{ mg/m}^3$ .

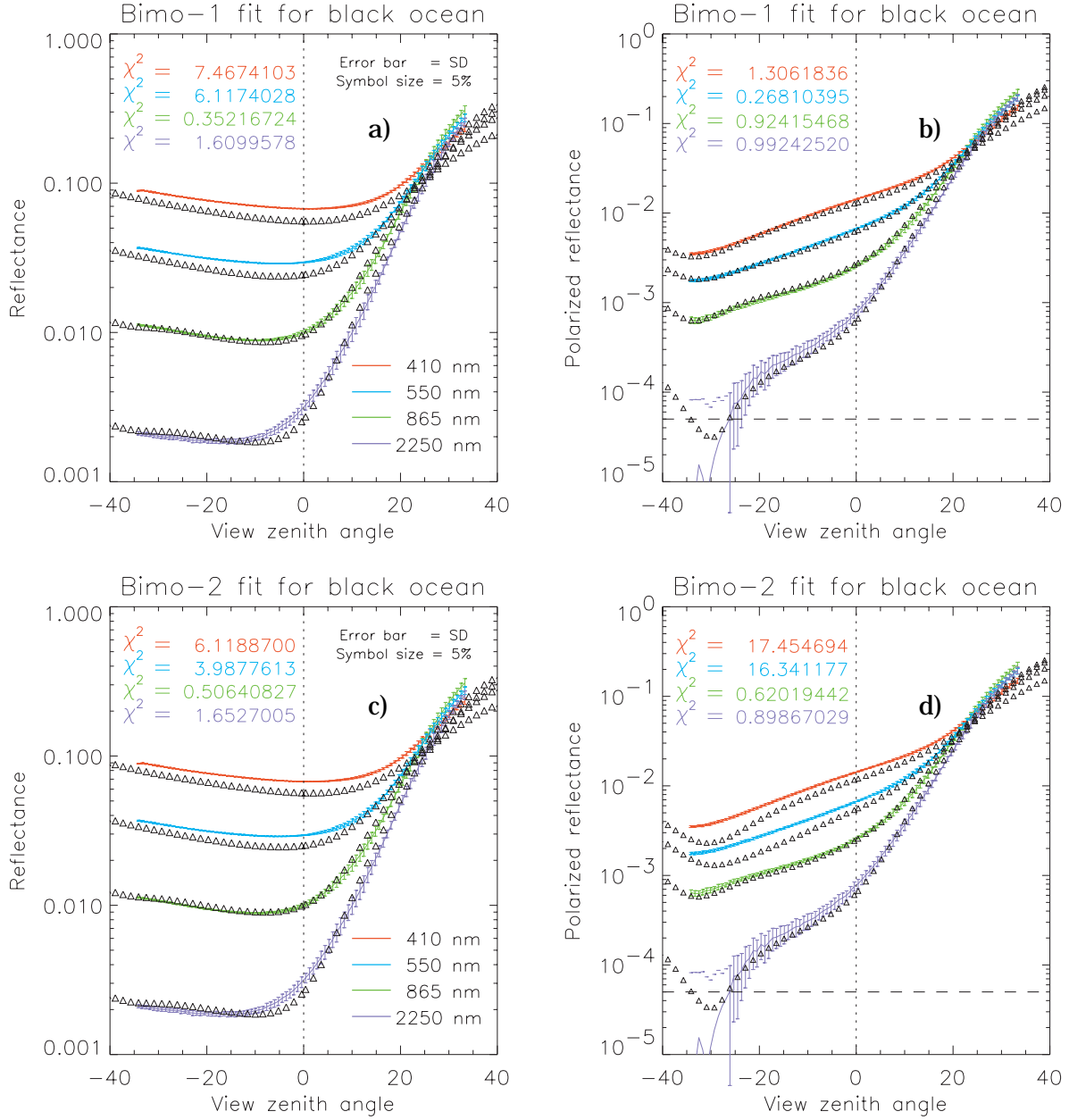


Figure 4. *a.* The fit (triangles) to the mean RSP reflectance data (lines) for  $\lambda = 0.410 \mu\text{m}$ ,  $0.550 \mu\text{m}$ ,  $0.865 \mu\text{m}$ , and  $2.250 \mu\text{m}$  using the accumulation and coarse mode components of the bimo-1 aerosol model listed in Table 2 and assuming a black ocean body. Error bars denote the standard deviation of the RSP data, and the size of the triangles represents a 5% uncertainty in the results of our simulations. The goodness of the fit is given for each wavelength by the normalized chi-square value in the upper left corner. *b.* Same as Fig. 4*a* except for polarized reflectances. *c.* Same as Fig. 4*a* except for the bimo-2 aerosol model listed in Table 2. *d.* Same as Fig. 4*b* except for the bimo-2 aerosol model listed in Table 2.



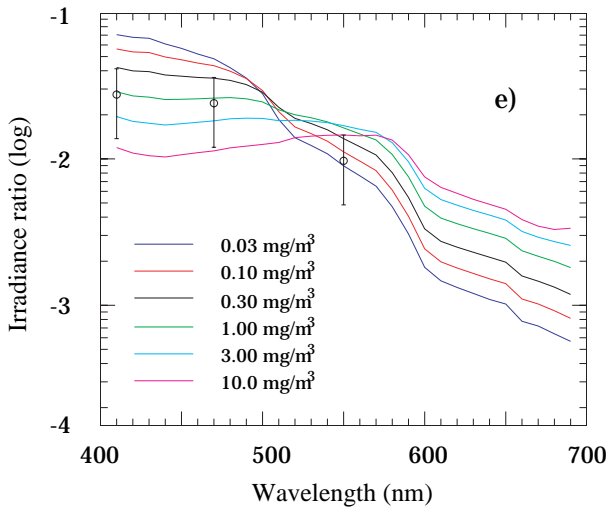
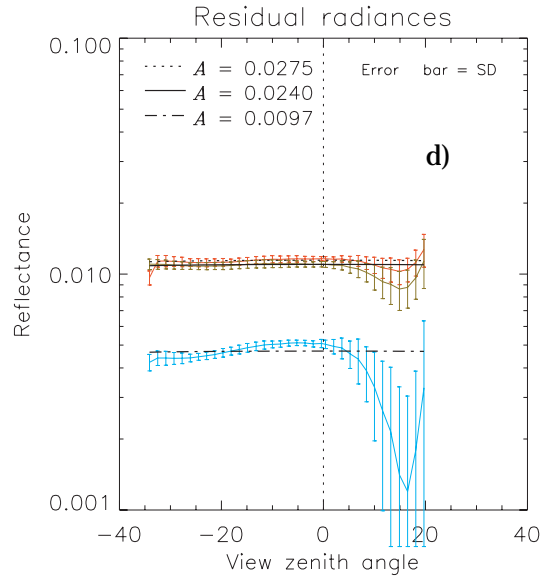
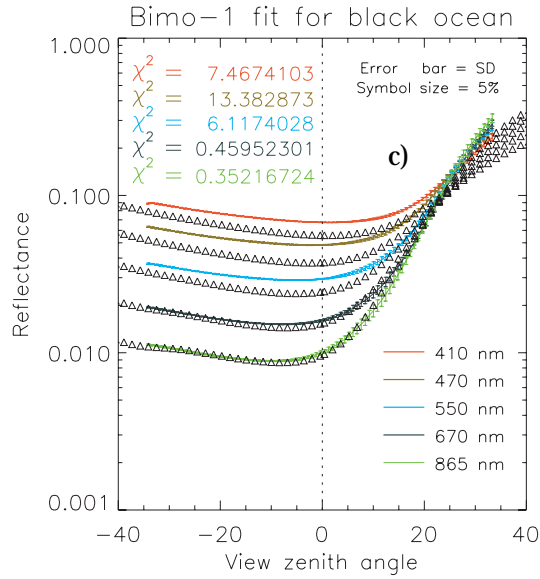
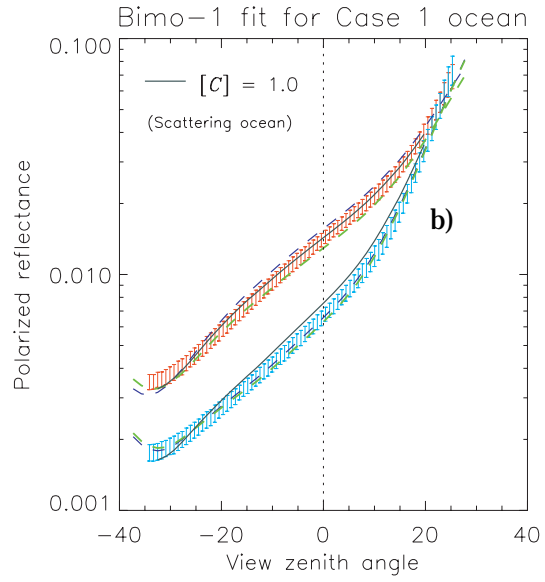
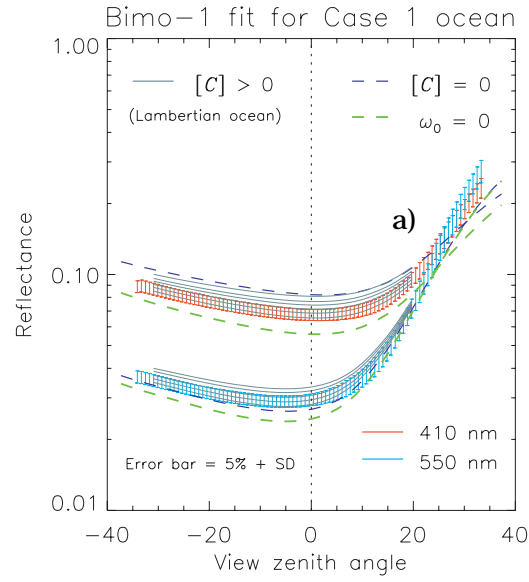


Figure 5. *a.* Error bars denote the sum of the standard deviation of the RSP data and a 5% uncertainty in model simulations, and are centered around the mean RSP data obtained at  $\lambda = 0.410 \mu\text{m}$  and  $0.550 \mu\text{m}$ . The green dashed lines show the corresponding bimo-1 aerosol model fits for a black ocean body (*cf.* Fig. 4*a*). The dark blue dashed lines are for including a molecular ocean, and the solid grey lines for using  $[C] = 0.01, 0.05, 0.10, 0.40,$  and  $1.00 \text{ mg/m}^3$  for  $\lambda = 410 \mu\text{m}$  and  $[C] = 0.01, 0.05, 0.15, 0.40,$  and  $1.00 \text{ mg/m}^3$  for  $\lambda = 550 \mu\text{m}$ . *b.* Same as Fig. 5*a* except for polarized reflectances and taking  $[C] = 1.00 \text{ mg/m}^3$  for the solid grey line. *c.* Lines and error bars show the mean RSP reflectance data and their standard deviation, respectively, for  $\lambda = 0.410 \mu\text{m}, 0.470 \mu\text{m}, 0.550 \mu\text{m}, 0.670 \mu\text{m},$  and  $0.865 \mu\text{m}$ . The symbols denote the corresponding fits using the bimo-1 aerosol model of Table 2 and assuming a black ocean body as in Fig. 4*a*. *d.* Lines are for the same wavelengths as in Fig. 5*c* but denote now the difference between the RSP data and corresponding fits. The black broken lines show the reflectances of water-leaving radiances simulated for an ocean body that is approximated by a Lambertian surface with albedo  $A$ . *e.* Irradiance ratios just below the ocean surface as a function of the wavelength and of  $[C]$ . The semi-empirical curves are reproduced from Morel (1988). Inserted are the ocean albedos  $A$  from Fig. 5*d* with error bars denoting the uncertainty (50%) of Morel's (1988) equations for irradiance ratios.

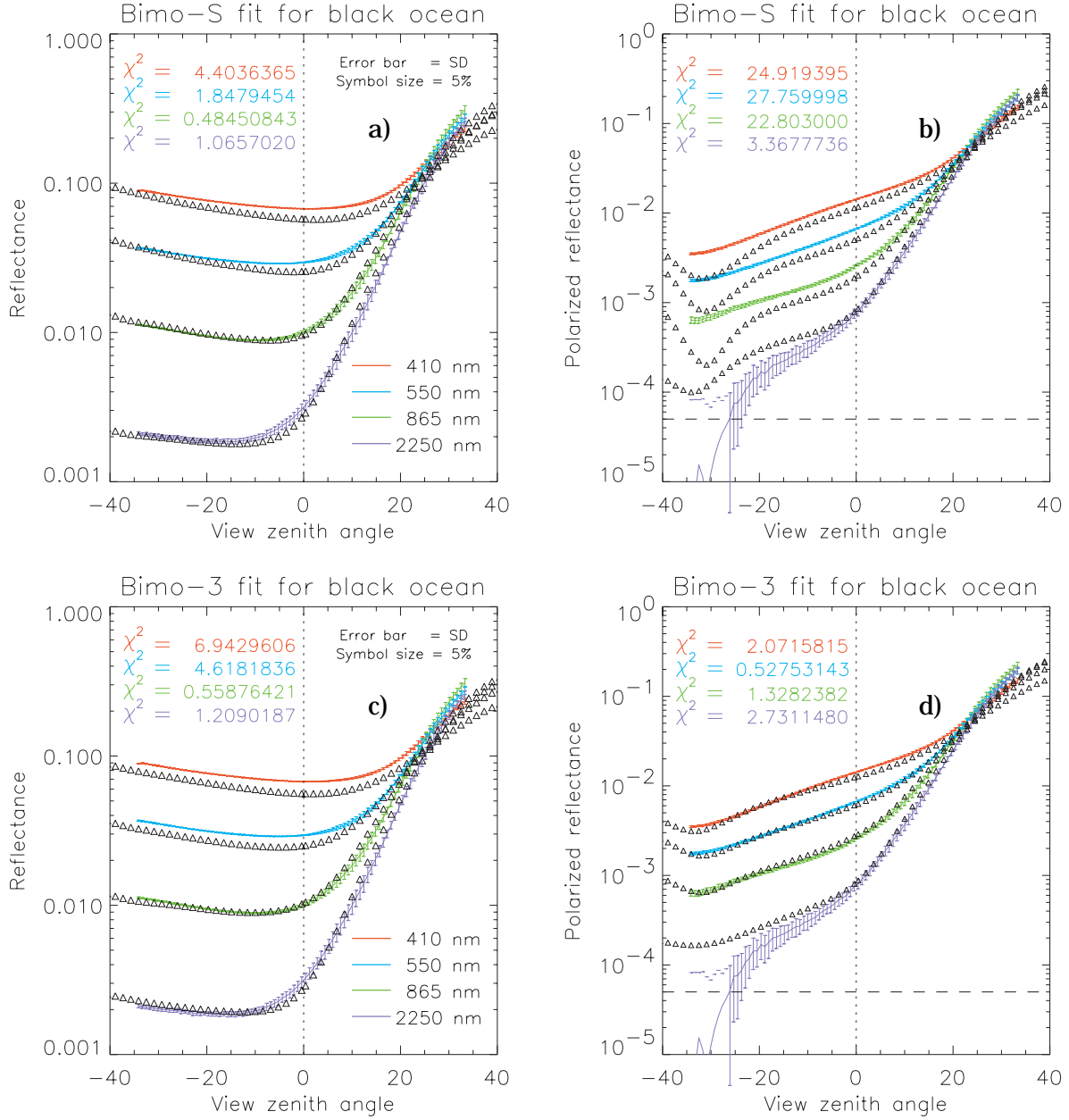


Figure 6. *a.* Lines and error bars show the mean RSP reflectance data and their standard deviation, respectively, for  $\lambda = 0.410 \mu\text{m}$ ,  $0.550 \mu\text{m}$ ,  $0.865 \mu\text{m}$ , and  $2.250 \mu\text{m}$  as in Fig. 4*a*. Symbols denote the corresponding fits using the bimo-S aerosol model specified in Table 3, and assuming a black ocean body. *b.* Same as Fig. 6*a* except for polarized reflectances. *c.* Same as Fig. 6*a* except for using the accumulation and coarse modes of the bimo-3 aerosol model listed in Table 2. *d.* Same as Fig. 6*c* except for polarized reflectances.

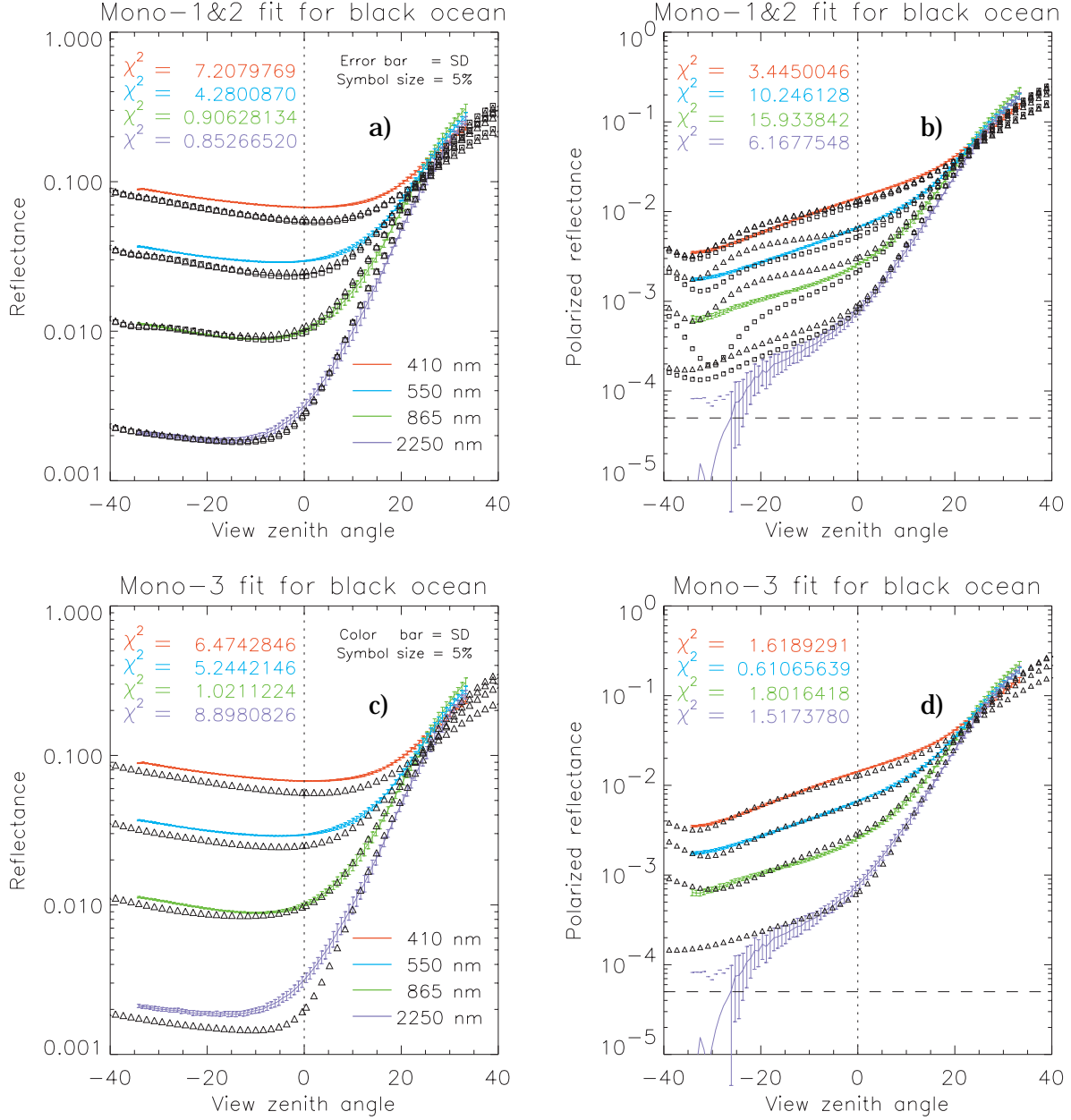


Figure 7. *a*. Lines and error bars show the mean RSP reflectance data and their standard deviation, respectively, for  $\lambda = 0.410 \mu\text{m}$ ,  $0.550 \mu\text{m}$ ,  $0.865 \mu\text{m}$ , and  $2.250 \mu\text{m}$  as in Fig. 4*a*. Triangles and boxes denote the corresponding fits using the mono-1 and mono-2 aerosol models listed in Table 2, respectively. The normalized chi-square values are given for the mono-1 fit. *b*. Same as Fig. 7*a* except for polarized reflectances. *c*. Same as Fig. 7*a* except for triangles denoting the fit using the mono-3 aerosol model listed in Table 2. *d*. Same as Fig. 7*c* except for polarized reflectances.

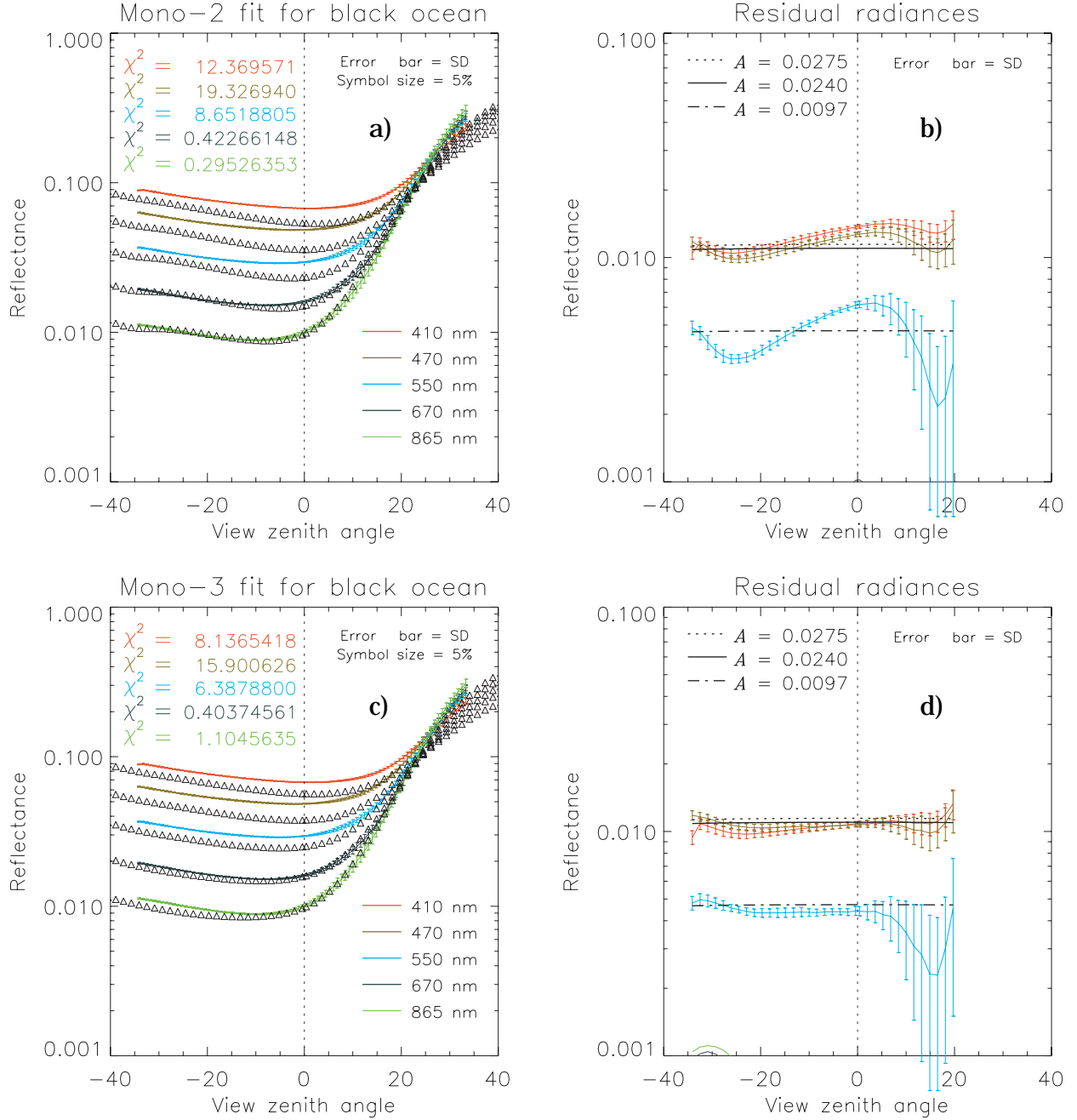


Figure 8. *a*. Lines and error bars show the mean RSP reflectance data and their standard deviation, respectively, for  $\lambda = 0.410 \mu\text{m}$ ,  $0.470 \mu\text{m}$ ,  $0.550 \mu\text{m}$ ,  $0.670 \mu\text{m}$ , and  $0.865 \mu\text{m}$  as in Fig. 5c. The symbols denote the corresponding fits using the mono-2 aerosol model listed in Table 2 and assuming a black ocean body. *b*. Lines are for the same wavelengths as in Fig. 8a but denote now the difference between the RSP data and corresponding fits. The black broken lines show the reflectances of water-leaving radiances simulated previously for the bimo-1 aerosol model case (*cf.* Fig. 5c). *c*. Same as Fig. 8a except for using the mono-3 aerosol model listed in Table 2. *d*. Same as Fig. 8b except for using the mono-3 aerosol model listed in Table 2

## Tables

**Table 1.** Comparison of capabilities of five instruments for tropospheric aerosol remote sensing.

Instrument	Number of bands	Spectral <sup>a)</sup> range	Number of view angles	Type <sup>b)</sup> of data
AVHRR	2	0.650 to 0.850	1	I
MODIS	7	0.470 to 2.130	1	I
MISR	4	0.443 to 0.865	9	I
POLDER	4 <sup>c)</sup>	0.443 to 0.865	<14	I,P
EOSP	9 <sup>d)</sup>	0.410 to 2.250	152	I,P

a) in  $\mu\text{m}$ ; b) I = Intensity, P = Polarization; c) 3 for P; d) 7 for P.

**Table 2.** Aerosol size parameters, refractive indices, optical thicknesses, and column number densities retrieved from RSP data (sub)sets.

Aerosol	$r_{\text{eff}}$ <sup>a)</sup>	$V_{\text{eff}}$	$m_{2250}$	$m_{550}$	$\tau_{550}$	$N$ <sup>b)</sup>
bimo-1 {	1.0	1.0	1.40	1.42	0.022	$2.2 \times 10^{10}$
	.25	.15	1.29	1.33	0.126	$6.2 \times 10^{11}$
bimo-2 {	1.0	1.0	1.40	1.42	0.022	$2.2 \times 10^{10}$
	.25	.15	1.34	1.40	0.121	$4.5 \times 10^{11}$
bimo-3 {	0.8	0.3	1.35	1.37	0.024	$9.4 \times 10^9$
	.30	.20	1.33	1.35	0.139	$4.0 \times 10^{11}$
mono-1	0.4	1.0	1.29	1.33	0.163	$1.4 \times 10^{12}$
mono-2	0.4	1.0	1.35	1.39	0.118	$9.0 \times 10^{11}$
mono-3	0.3	0.3	1.31	1.35	0.169	$6.5 \times 10^{11}$

a) in  $\mu\text{m}$ ; b) in  $\text{m}^{-2}$ .

**Table 3.** Specification for the bimo-S model, and the optical thicknesses retrieved from the RSP total reflectance data set

Aerosol	$r_c$ <sup>a)</sup>	$\sigma$	$m_{2250}$	$m_{865}$	$m_{550}$	$m_{410}$	$\tau_{550}$
oceanic	0.30	2.00	1.31	1.35	1.35	1.36	0.048
soluble	.005	2.99	1.42	1.52	1.53	1.53	0.067

a) in  $\mu\text{m}$